

*Original Research*

# Coupling Microbial Fuel Cells with Electrocoagulation Cells to form an Integrated System for Wastewater Treatment

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*Received: 7 January 2018*

*Accepted: 26 March 2018*

## Abstract

Microbial fuel cells (MFCs) and electrocoagulation cells (ECCs) are two emerging technologies in the treatment of wastewater. The integration between MFCs and ECCs has not been reported yet. This work studied the ability to couple MFCs with an ECC to form an integrated system for wastewater treatment. Two types of wastewater were examined: synthetic wastewater containing a mixture of glucose and soluble starch, and real municipal wastewater. A series of MFCs could provide sufficient energy for the electrocoagulation process. The results showed that the removal efficiencies of COD, TDS, and TSS were 95.4%, 88.4%, and 93.8%, respectively, for synthetic wastewater, while these values were 83.7%, 57.5%, and 85.8%, respectively, for real wastewater. The energy harvested from the MFCs to ECCs when using synthetic wastewater was more than that harvested using real wastewater. The capital cost of the integrated system is high using MFCs and ECCs, but it will significantly reduce the operational cost compared to ECCs.

**Keywords:** microbial fuel cells, electrocoagulation, wastewater treatment, integrated treatment system, energy storage device

## Introduction

Cost is considered the main factor that governs the choice of one wastewater treatment alternative over another. Energy is one of the parameters that raises or lowers the total cost of a treatment system. Based on this concept, many researchers all over the world have started to focus on low-cost wastewater treatment systems, and systems that consume less energy or even those that can generate energy [1-4]. One of the promising technologies

that can generate energy is the microbial fuel cell (MFC). MFCs are systems that can produce electric current through microorganisms [5-7]. These systems can be considered voltaic cells. MFCs can be designed to be of single compartment or dual compartment [8]. An MFC consists of two anodes: one anaerobic anode and one aerobic cathode, which are separated by a proton exchange membrane. The membrane allows only protons or other cations to transfer from the anodic compartment to the cathode compartment [8]. A wire is used to connect the two electrodes to close the external circuit. The substrates are oxidized in the anode compartment via microorganisms [9]. By

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removing the proton-permeable membrane, the cost of an MFC unit could be decreased [10, 11]. The current research on MFCs is still at its beginning and further efforts are needed to meet the challenges before wide application is possible. The main challenge is the power output of MFCs, which is still below desired levels [9]. An MFC can be used to operate an electronic device if the generated energy can be stored in an external storage device, such as a capacitor or rechargeable battery. Then the stored energy can be used when needed [12].

Another type of treatment system, which has a similar configuration as the MFCs, is the electrocoagulation cell (ECC). These systems can be considered electrolytic cells in which an external power supply must exist. Electrocoagulation is a promising treatment method that combines the advantages of conventional coagulation and flotation [13]. Dissolution of the electrode material is the main concept used in the electrocoagulation process [14]. Several electrodes can be used, including aluminum, iron, copper, and other electrodes [15]. In ECC, aluminum is dissolved from the anode into the reaction solution that interacts with the hydroxyl ions produced at the cathode to form aluminum hydroxide. The production of aluminum hydroxides can remove contaminants from water and wastewater [16]. An electrolyte, such as sodium chloride (NaCl), is always used to increase the conductivity of wastewater to be treated by an electrochemical process and decreases the passivation of the aluminum surface to promote electrocoagulation efficiency [17, 18]. The addition of NaCl can also reduce the electrical energy consumption of electrocoagulation, as it increases the conductivity of the wastewater. Previous studies showed that electrocoagulation was strongly enhanced when using aluminum electrodes due to the formation of dispersed aluminum-hydroxide complexes through hydrolysis of the aluminate ion [17]. ECC showed good results in the treatment of different types of wastewater over different values of external voltage.

In order to harvest energy from MFCs, capacitors can be used to capture and store energy and increase voltage for practical use [19]. The rechargeable battery could extract more energy compared to an ultracapacitor [20]. To the best of our knowledge, integration between MFCs and ECCs has not yet been reported. Although previous research has shown that MFCs produce power

densities of small values [19], they may provide a suitable current density for ECCs. The main objective of this study was to investigate the feasibility of coupling both MFCs and ECCs into one integrated system to use the energy produced from one system to feed the other, and thus significantly decrease the total cost of the treatment process.

## Materials and Methods

### Characteristics of Wastewater

Two types of wastewaters were used. First, synthetic wastewater consists of the following (per liter of distilled water): 0.2 g  $\text{NH}_4\text{Cl}$ , 0.15 g  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , 0.33 g KCl, 0.50 g NaCl, 3.15 g  $\text{MgCl}_2$ , 1.26 g  $\text{K}_2\text{HPO}_4$ , 0.42 g  $\text{KH}_2\text{PO}_4$ , and trace metals (1 mL) [21]. A mixture of glucose and soluble starch were used as a carbon source. The value of TDS was high in order to increase conductivity to enhance the performance of the electrocoagulation process. After that, real municipal wastewater, obtained from the Zenin wastewater treatment plant in Egypt, was used in the integrated system. Table 1 shows the characteristics of synthetic wastewater and real wastewater used in this study.

### System Setup and Operating Procedures

Treatment of synthetic wastewater was examined using both single-chamber MFC and ECC. The MFC used in the experiments had a circular chamber, and its effective volume was 250 ml. Graphite fiber felt electrodes were used for both the anode and cathode in the MFC (MudWatt, USA), and the vertical distance was adjusted to be 5 cm. To facilitate the startup, the anode electrodes were obtained from other working microbial fuel cells. The total surface area of each electrode was 130  $\text{cm}^2$ . The MFC was operated in batch mode, and the cycle length of each experiment was 72 hours [8, 11]. Before starting the experiments, nitrogen gas was used to flush the compartment of any air. The ECC used in the experiments had a circular chamber of an effective volume of half liter. The anode electrode was made of an aluminum sheet, while the cathode electrode was made of a stainless-steel sheet. The total surface area of each electrode was 60  $\text{cm}^2$ , and the horizontal distance between them was 3 cm. The ECC was operated in batch mode, and the cycle length of each experiment was 60 minutes. The integrated system was formed of six MFCs, placed in series, followed by one ECC, as shown in Fig. 1. The electricity generated from MFCs was stored in two rechargeable batteries each of 1.2 V and 800 mAh. Then these batteries were used in a series to provide electricity to the ECC. The batteries were completely discharged before using them in the experiments. The wastewater used was maintained under mixing at a rate of 100 rpm during the operation of both MFCs and the ECC.

Table 1. Characteristics of wastewater.

Parameter	Synthetic wastewater	Real wastewater
pH	7	7.18
COD	1000 mg/l	467 mg/l
TDS	1000 mg/l	537 mg/l
TSS	580 mg/l	207 mg/l

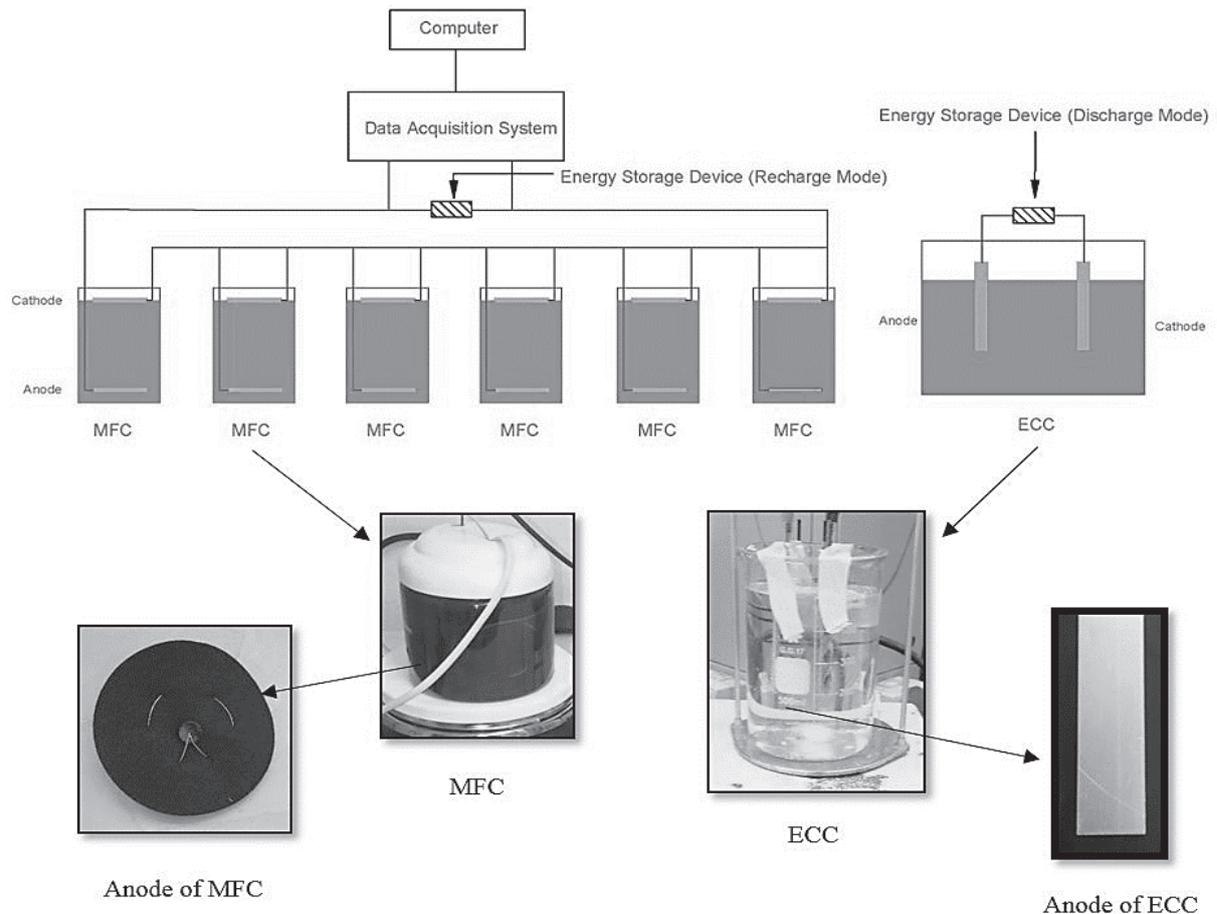


Fig. 1. Schematic diagram of integrated system of MFCs and ECC.

### Analysis

Samples were taken using a sterile syringe. The COD concentration was measured using HACH high range (0-1500 mg/l) COD vials and DR 220 spectroscopy (HACH, USA). To decrease the resistance of the system, titanium wires were used to connect the cathode and anode electrodes. The voltage generated across the external resistance of the six MFCs was measured at one-hour intervals using a data acquisition system (USB DrDAQ Data Logger, Pico Tech., UK) connected to a computer. The pH and TDS at the start and end of the experiments were measured using a multiparameter meter (Hanna HI 9813-6 N, USA). The coulombic efficiency was calculated using the following equation:  $CE = \frac{8 \int_0^t Idt}{FV\Delta COD}$ , where  $\int_0^t Idt$  is the actual coulombs generated over the time period (t), F is the Faraday constant (96500 C/mol electrons), V is the active volume of the chamber, and  $\Delta COD$  is the amount of COD removed over a time period (t) [22]. The morphology of electrodes was examined using an atomic force microscope (AFM) and a scanning electron microscope (SEM). Experiments were done under a constant temperature of  $25 \pm 0.5^\circ\text{C}$ .

### Results and Discussion

#### Performance of MFC for Synthetic Wastewater Treatment

Synthetic wastewater was treated using a single-chamber microbial fuel cell. Fig. 2a) shows the removal of COD over a hydraulic retention time of 72 hours. The removal efficiency increased with time, reaching the maximum value of 43.4% at the end of the cycle length. The rate of removal was almost constant over time due to the use of the inocula previously adapted to the MFCs and the biodegradability of the substrate. The coulombic efficiency was found to be 77.4%, which means that most removal occurred due to electrogenic bacteria.

#### Performance of Electrocoagulation Cell for Synthetic Wastewater Treatment

Synthetic wastewater was treated using an ECC. Fig. 2b) shows the removal of COD over the hydraulic retention time of 60 minutes. The removal efficiency increased with time, reaching the maximum value of 85.4% after 60 minutes. The rate of removal was

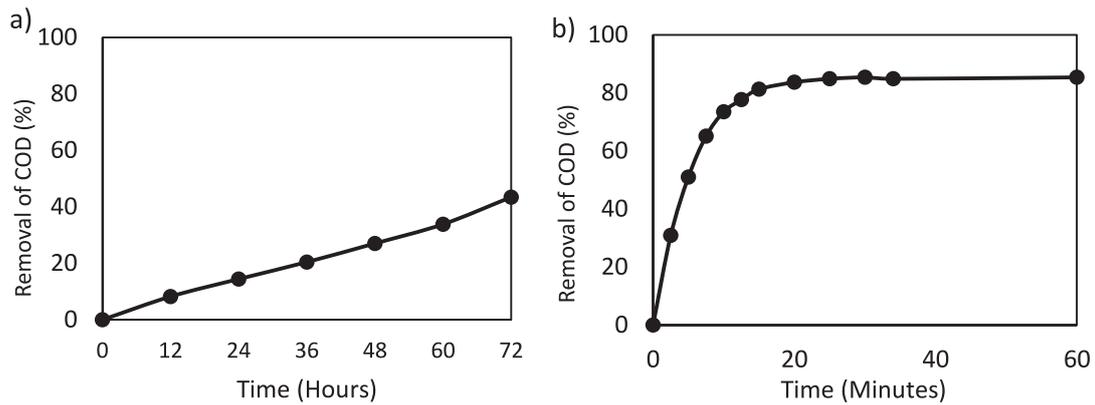


Fig. 2. Efficiency of COD removal for synthetic wastewater: a) using MFC and b) using ECC.

very high at the beginning of the experiment until 15 minutes, after which the removal rate dramatically decreased. This may be due to the reduction in conductivity, which in turn may increase the internal resistance of the system and thus decrease the efficiency of the electrocoagulation process.

#### Performance of MFCs Followed by an ECC for Wastewater Treatment

The integrated system of MFCs and an ECC was used for treating synthetic wastewater. The time for treatment was 72 hours in MFCs, followed by 60 minutes in ECC. Fig. 3a) shows the voltage generation over time. The voltage of the series of MFCs increased with time, reaching its peak value of 3.3 V after about 60 hours and then began to decrease until the end of the cycle length. The decrease in voltage generation was due to substrate consumption during the treatment process. The value of the voltage was above 2.4 V after 36 hours until the end of the cycle time. During this period, a storage device (two batteries each of 1.2 V) was recharged to be used to operate the ECC at 2.4 V for 60 minutes. Fig. 3b) shows the values of the influent and effluent of COD, TDS, and TSS for the

synthetic wastewater when using the integrated system. The removal efficiencies of COD, TDS, and TSS were 95.4%, 88.4%, and 93.8%, respectively. The value of the COD removal was higher than that obtained when using the MFCs and ECC separately. This is because coupling both MFCs and the ECC into one system led to a decrease in the influent value of COD to ECC, which in turn significantly decreased the value of effluent COD compared to previously mentioned results. The reduction in TDS was mainly due to the removal mechanism in ECC, which depends on the movement of ions between the anode and cathode. The removal of TSS took place through two mechanisms: a) biodegradation inside MFCs and b) coagulation in ECC.

Based on the success of the integrated system in the treatment of synthetic wastewater, the system was then investigated to determine its ability to treat real wastewater. The time for treatment was 72 hours in the MFCs followed by 60 minutes in the ECC. Fig. 4a) shows the voltage generation over time. The voltage of the series of MFCs increased with time until reaching its peak value of 1.86 V after about 60 hours, then began to decrease until the end of the cycle. The decrease in voltage generation was due to substrate consumption during the treatment process. The value of the voltage

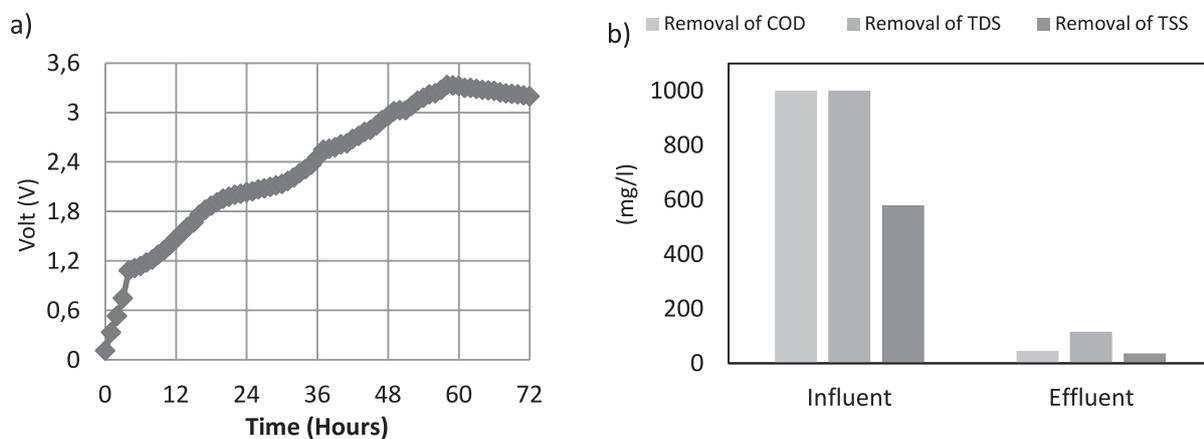


Fig. 3: a) Voltage generation from MFCs over time during treatment of synthetic wastewater and b) Influent and effluent COD, TDS, and TSS of synthetic wastewater.

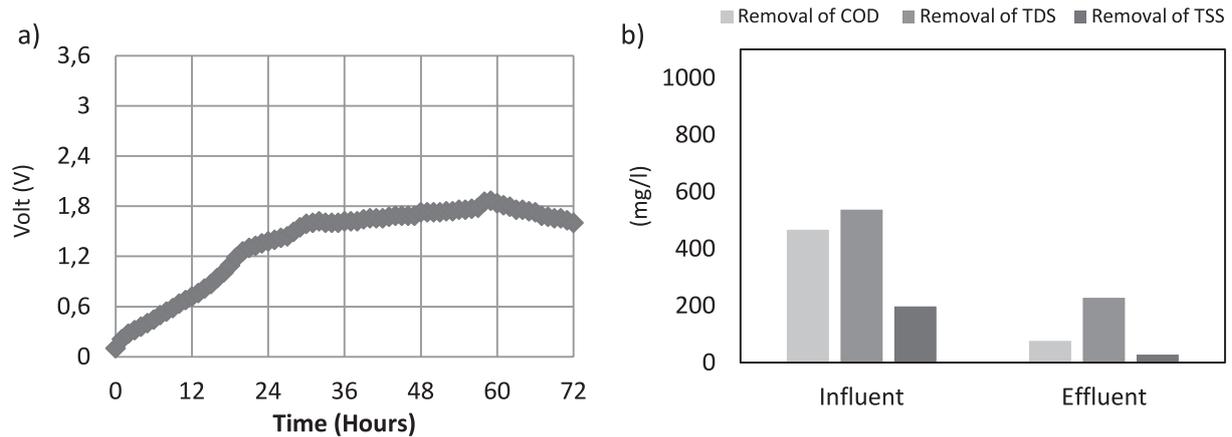


Fig. 4: a) Voltage generation from MFCs over time during treatment of real wastewater and b) Influent and effluent COD, TDS, and TSS of real wastewater.

was above 1.2 V after 20 hours until the end of the cycle time. During this period, the voltage did not exceed 2.4 V, which means that this system will not be able to recharge the series batteries. Thus, a storage device (1.2 V) was recharged to be used to operate the ECC at 1.2 V for 60 minutes. The values of the voltage generated were less than those obtained when treating the synthetic wastewater due to the complexity of the real wastewater, which in turn can encourage biodegradation through non-electrogenic bacteria. This was proved by operating one MFC to treat real wastewater, and by the end of the cycle time, the coulombic efficiency was found to be around 60%, which is less than that obtained when treating synthetic wastewater under the same conditions. Additionally, the value of influent COD in real wastewater was less than that for synthetic wastewater. Fig. 4b) shows the values of the influent and effluent of COD, TDS, and TSS for real wastewater. The removal efficiencies of COD, TDS, and TSS were 83.7%, 57.5%, and 85.8%, respectively. The value of COD removal was lower than that obtained when treating synthetic wastewater. This is because the applied voltage in the electrocoagulation process is about one half that applied during the treatment of the synthetic wastewater, which significantly affected the efficiency of the process. The reduction in TDS was mainly due to the mechanism of removal in the ECC, which depends on the movement of ions between the anode and cathode. The reduction in TSS was due to the biodegradation process in the MFCs. Then the coagulation process occurred in the ECC, which resulted in solids removal through different mechanisms, such as enmeshment in a precipitate. The pH increased from 7.1 to 7.5 because of the continuous hydroxyl ion production that occurs at the cathode. It is worth noting that, at a neutral pH, the production of hydrogen bubbles produced at the cathode is the smallest and finest; thus, sufficient surface area for gas–liquid–solid interfaces and mixing efficiency is provided, which enhances the aggregation of the tiny destabilized particles and colloids [17].

### Morphology of Anode Electrodes

The surface of the anode electrode of one of the MFCs was examined using an AFM before and after the treatment of the real wastewater, as shown in

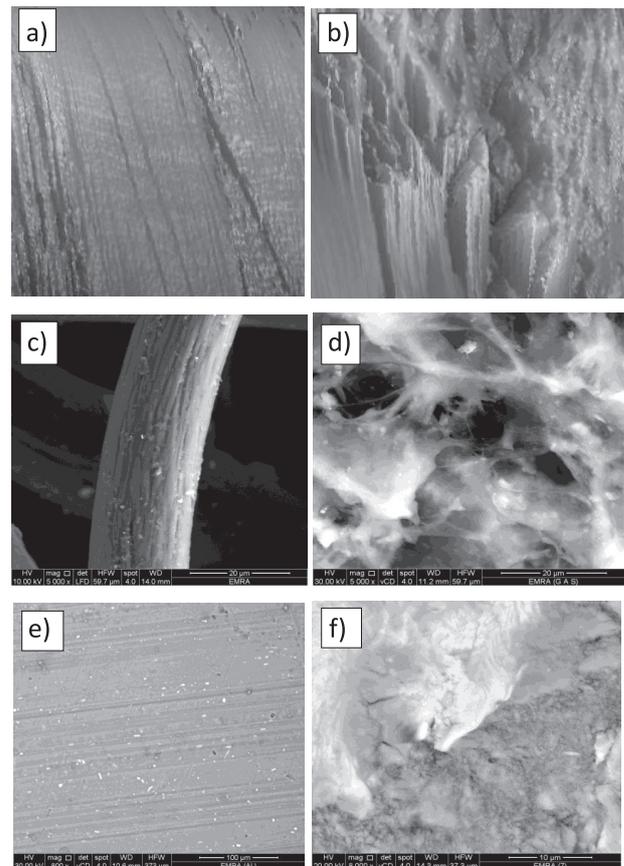


Fig. 5: a) AFM image of blank anode electrode used in the MFC, b) AFM image of anode electrode used in the MFCs after treatment, c) SEM image of blank anode electrode used in MFCs, d) SEM image of anode electrode used in MFCs after treatment, e) SEM image of blank aluminum electrode used in ECC, and f) SEM image of aluminum electrode used in ECC after treatment.

Table 2. Costs of different systems for treatment of 1 m<sup>3</sup> per year.

System	Cost of electrodes (\$/m <sup>2</sup> )	Total area of electrodes required for 1 m <sup>3</sup> of wastewater	Total capital cost (\$/m <sup>3</sup> )	Cost of electricity required per year for 1 m <sup>3</sup> of wastewater (\$/m <sup>3</sup> )	Cost of electrode replacement per year (\$/m <sup>3</sup> )	Total operating cost per year (\$/m <sup>3</sup> )
MFC	60	104 m <sup>2</sup>	6240	-	-	-
ECC	40	24 m <sup>2</sup>	960	3363.84	2880	5643.84
Integrated system	-	-	7200	-	2880	3600

Fig. 5a) and 5b). Before operation, the anode surface was less corrugated, while after the end of the treatment process in the MFC, the surface was much more corrugated. This is due to the increasing in mass of attached microorganisms to the anode surface during the operation, which conformed to the power generation. This change was confirmed through SEM images of electrodes before and after treatment as shown in Fig. 5c) and 5d). Since chemical changes occurred in the surface of the electrodes in the electrocoagulation process, the morphology of the aluminum electrode used in the ECC was examined using an SEM before and after the treatment of the real wastewater to compare the surface texture, as shown in Fig. 5e) and 5f). The images show that the surface of the aluminum electrode contained only a few dents, while the surface became rougher at the end of the experiments. This change in surface was due to the anode consumption through electrode dissolution to form aluminum hydroxides.

### Cost Analysis

The total cost of any system includes two main components, which are the capital cost and operation and maintenance costs. For MFCs, the main part of the cost is the cost of the electrodes, which can be considered a capital cost, while the operational cost can include the maintenance of the electrodes in case of fouling. In the ECC, the capital cost includes the construction cost, which primarily includes the electrodes, while the operating cost includes both the cost of the electricity needed and the replacement of the electrodes due to the depletion of scarified metal. To obtain a rough estimate of cost, the author will consider the cost of removal of COD from 1 m<sup>3</sup> of synthetic wastewater based on the previously mentioned results. Table 2 shows the estimated cost for the MFC, ECC, and the integrated system. For simplicity, the capital cost will be considered the cost of the electrodes. The replacement of electrodes of the ECC can be assumed to occur three times per year, and the cost of electricity can be assumed to be 0.2 \$/kwh. The Power can be obtained by multiplication of voltage times the current intensity. The results in the table show that, although the capital cost of the integrated system could be more than that for the MFC and ECC, the main two benefits

of using the integrated system will be the reduction that would occur in the operation cost of the system in addition to the increase in removal efficiency of COD.

### Conclusion

This work presented the study of the ability to couple MFCs with an ECC to form an integrated system for wastewater treatment. Two types of wastewater were examined: synthetic wastewater containing a mixture of glucose and soluble starch, and real municipal wastewater. A series of MFCs could provide sufficient energy for the electrocoagulation process. The results showed that the removal efficiencies for COD, TDS, and TSS were 95.4%, 88.4%, and 93.8%, respectively, for the synthetic wastewater, while these values were 83.7%, 57.5%, and 85.8%, respectively, for the real wastewater. The energy harvested from the MFCs to the ECC when using synthetic wastewater was more than that using the real wastewater. Further studies for physical and hydraulic conditions of both MFCs and ECCs are needed to obtain the conditions for maximum energy generation and minimum cost.

### Acknowledgments

The author wishes to thank specialists at the Housing and Building National Research Center; at the Micro Analytical Center, Cairo University; and the Central Laboratory of Egyptian Mineral Resources Authority for help with analyses.

### Conflict of Interest

The authors declare no conflict of interest.

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